

# Electrodeposition for Low-Cost, Water-Based Electrode Manufacturing

Vehicles Technology Office  
2018 Annual Merit Review

Project ID: BAT263



**Stuart Hellring (PI)**  
**June 19, 2018**

**Contributors:**

PPG – Landon Oakes, Haley Orlor, Ryan Plazio

ANL - Andrew Jansen, Greg Krumdick, Ozge Kahvecioglu Feridun

ORNL - David Wood, Marissa Wood, Jianlin Li

Navitas – Mike Wixom, Pu Zhang

# Overview

## Timeline

- Project start date: January 1<sup>st</sup>, 2016
- Project end date: December 31<sup>st</sup>, 2018
- 66% complete

## Barriers

- High material processing cost
- High manufacturing cost
- Toxic material exposure

## Budget

- Total project funding:\$3,999,034
  - DOE share: \$1,399,275
  - FFRDC: \$1,600,000
  - Contractor share: \$999,759
- Project is fully funded.
- Funding for FY 2017: \$762,346
- Funding for FY 2018: \$826,415

## Partners

### Argonne National Lab

#### Project leads:

- Greg Krumdick (MERF)
- Andy Jansen (CAMP)

### Oak Ridge National Lab

Project lead: David Wood III

### Navitas System

Project lead: Mike Wixom



# 1. Relevance

## Overall Objectives

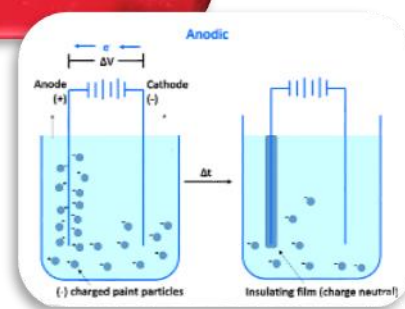
- Reduce electrode manufacturing cost using electrocoat processing.
- Improve the environmental friendliness with water-based battery processing.

## Objectives this Period

- Achieve target energy density by optimizing formulation and application.
- Achieve uniform films during simultaneous double-sided coating.
- Validate full-cell, pouch cell battery performance.
- Design and build pilot-scale roll-to-roll coater.
- Cost comparison of electrocoat to traditional processes.
- Improve battery performance using electrocoat.

## Impact

- Successful production of electrocoated cathodes will:
  - Reduce cell manufacturing cost.
  - Enable waterborne manufacturing.
  - Eliminate the need for using toxic solvents.
  - Facilitate automotive OEM and consumer acceptance of electric vehicles.
  - Allow for the creation of the next generation of US-based advanced battery manufacturing.




# Milestones

Date	Milestones and Go/No-go	Status
June 2017	<b>Milestone:</b> Formulation / application parameters are optimized sufficient to produce an electrode with an energy density of 2.5-3.0 mAh/cm <sup>2</sup>	Complete
December 2017	<b>Milestone:</b> Pouch cells > 0.2 Ah are tested	Complete
April 2018	<b>Milestone:</b> Mini-coater is designed, built, and prepared for operation.	Designed and Built Install - On track
December 2017	<b>Milestone:</b> BatPac model updated and adjusted cost estimate obtained	Complete
December 2017	<b>Go/No-go:</b> Demonstrate ability to produce kg quantities of the active material.	Complete
December 2017	<b>Go/No-go:</b> Electrodes will either have reached a loading density of 2.0 mAh/cm <sup>2</sup> or a clear path to achieve metric that will be identified.	Complete
July 2018	<b>Milestone:</b> Electrodes are produced on the mini-coater that can be used for cell deliverables.	On track
October 2018	<b>Milestone:</b> 12 baseline and 12 electrocoated cathodes will be evaluated in double layer pouch cells	On track
January 2019	<b>Milestone:</b> 35 electrocoat and 12 baseline prismatic cells >1 Ah will be assembled and tested. 18 optimized cells will be delivered to DOE for evaluation	On track
January 2019	<b>Milestone:</b> Root cause failure mechanisms identified	On track

# 1. Approach: Use Electrocoat to Overcome Current Process Barriers

## Eliminate Toxic Solvent Exposure Costs

**NMP Solvent  
GHS Hazards Label**



Signal Word: **WARNING!**  
Hazard Statements:  
H227 - Combustible liquid and vapor.  
H316 - Causes mild skin irritation.  
H320 - Causes eye irritation.  
H335 - May cause respiratory irritation.  
H360 - May damage fertility or the unborn child.

NFPA 704

Health	2
Flammability	2
Instability/Reactivity	0

**Water  
GHS Hazards Label**


**No GHS Warnings**

Health	0
Flammability	0
Physical Hazard	0
Personal Protection	X

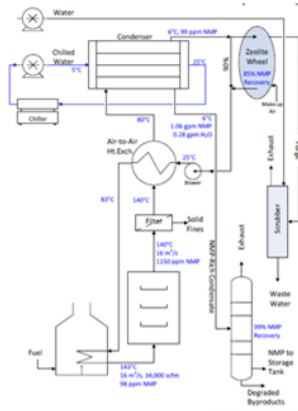
Health 0 Flammability 0 Instability/Reactivity 0

Integrated with other VTO W/B Projects at ORNL

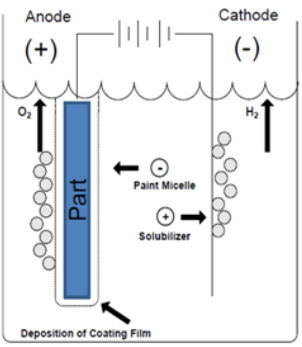
## Eliminate Costly Toxic Solvent Recovery



**NMP Recovery Process**  
Shabbir Ahmed, ANL  
2015 VTO AMR ES228



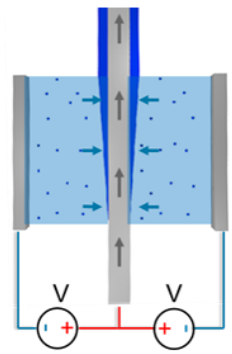
## Lower Drying Costs



Wet Film going into oven:

- High solids
- Low solvent
- Low VOC
- No LEL limitation

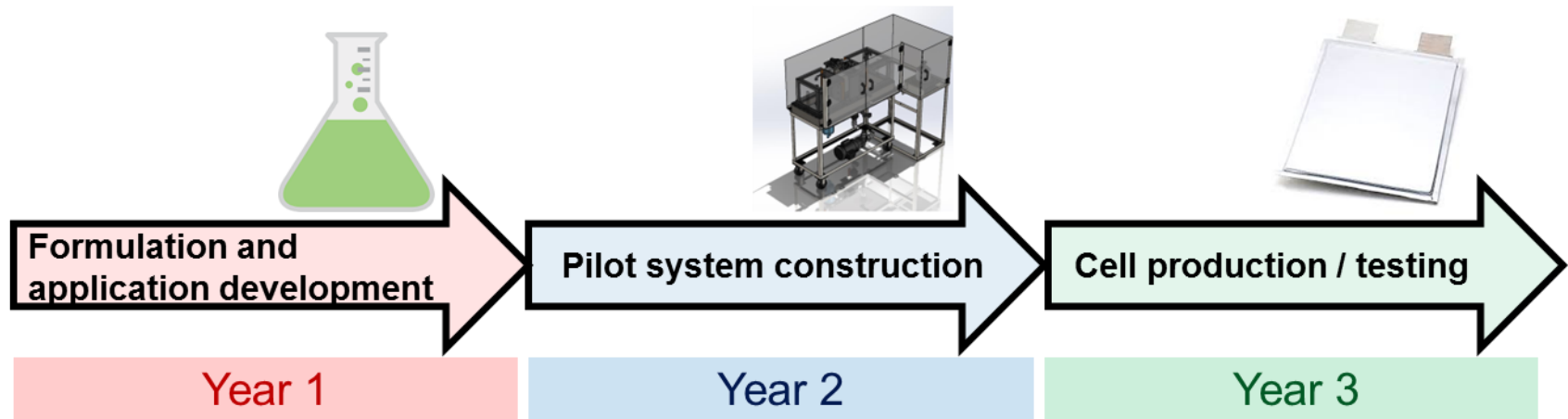
## Eliminate 2-Coat / 2-Cure Coating Process



- **Simultaneously** coat both sides
- One pass through oven
- Deposition controls uniformity
- Particle assembly controls porosity



## 2. Approach: Electrocoat System Design



### Formulation Development

- **Design, synthesize and screen electrocoat chemistries to accomplish:**
  - Stable electrocoat bath with suitable cathode compositions.
  - Composition coalesces into a cathode coating on the current collector.
  - Bench-top, batch scale electrocoat process optimization
  - Robust cathode coating for out-of-cell manipulation.
  - Outstanding performance when operated as a cathode.

### Pilot System Design and Production

- Design, build and install a pilot scale roll-to-roll electrocoat system.
- Develop water-based continuous roll-to-roll electrocoat process to demonstrate production of rapid, double-sided coating of uniform cathodes with high areal capacities.
- Cost analysis of electrocoat manufacturing based on pilot operation data.

### Cell Production / Testing

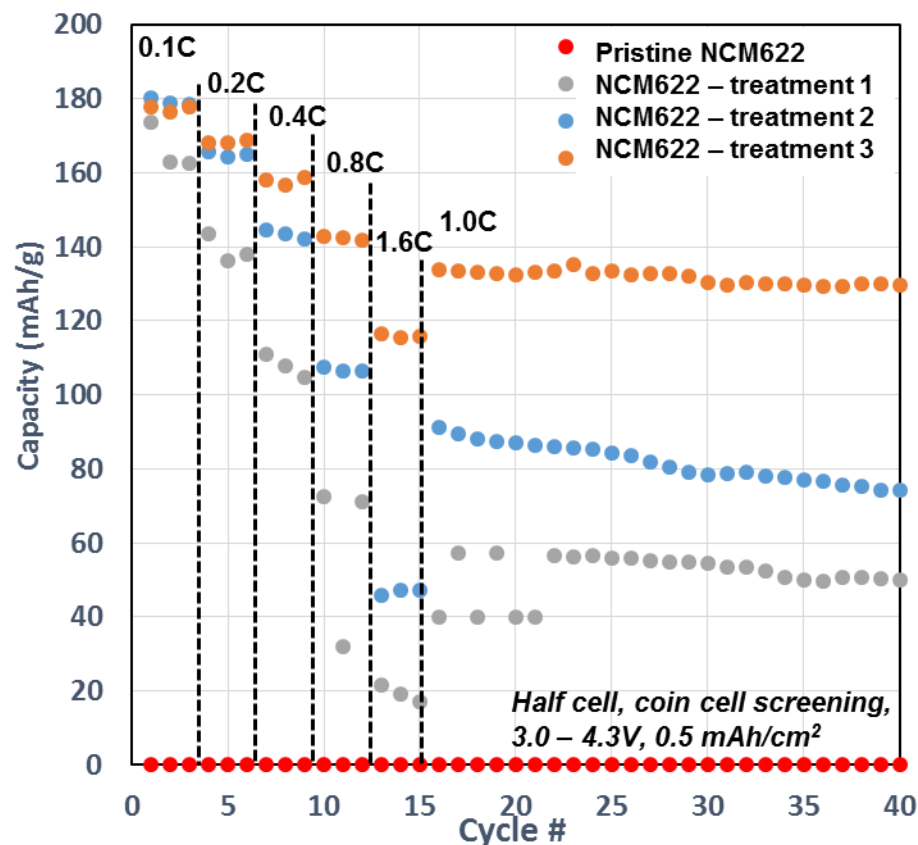
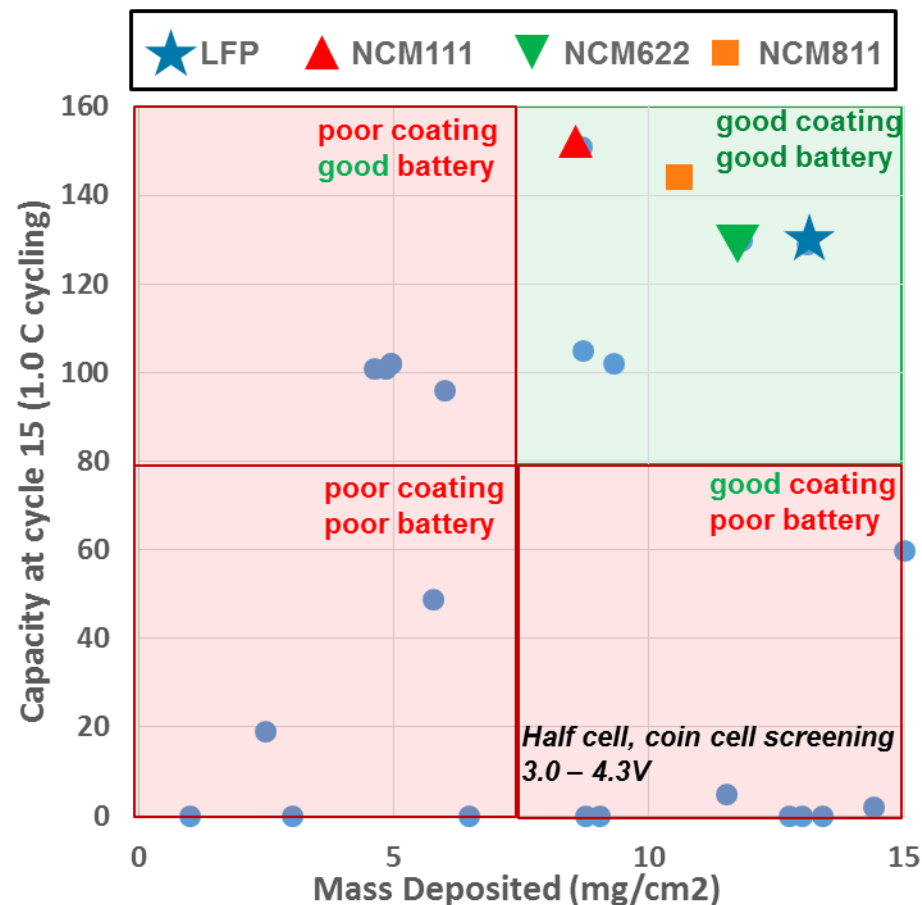
- Manufacture prismatic pouch cells using electrodes coated on the pilot scale coater



# 1. Technical Accomplishments: Commercial Material Screening

Over 20 commercial and custom-synthesized materials screened for stability toward electrocoat.

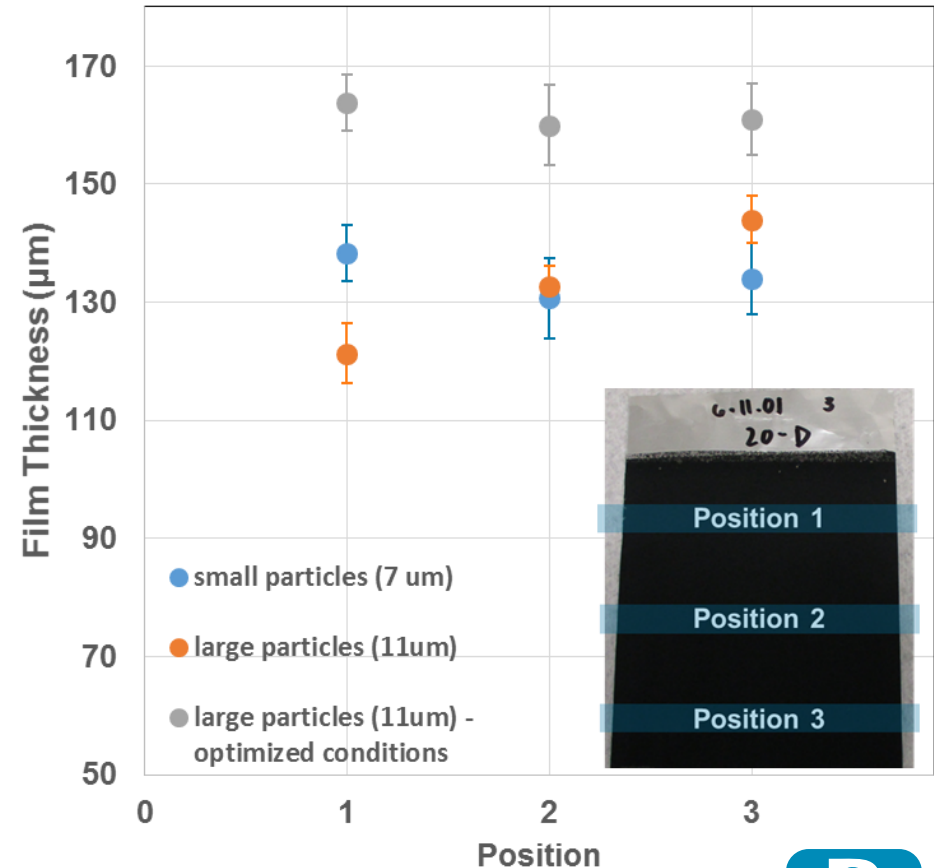
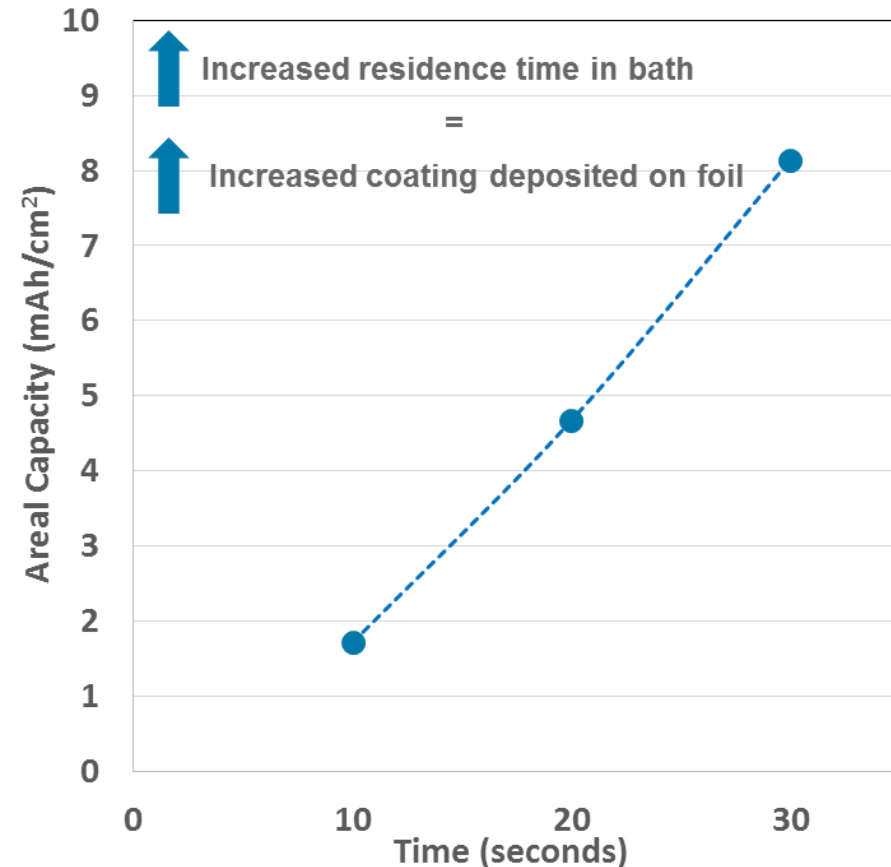
Surface treatments can stabilize high-nickel active materials toward electrocoat.



## 2. Technical Accomplishments: Coating System Development

Uniform areal densities up to 8.2 mAh/cm<sup>2</sup> are reliably fabricated using electrocoat.

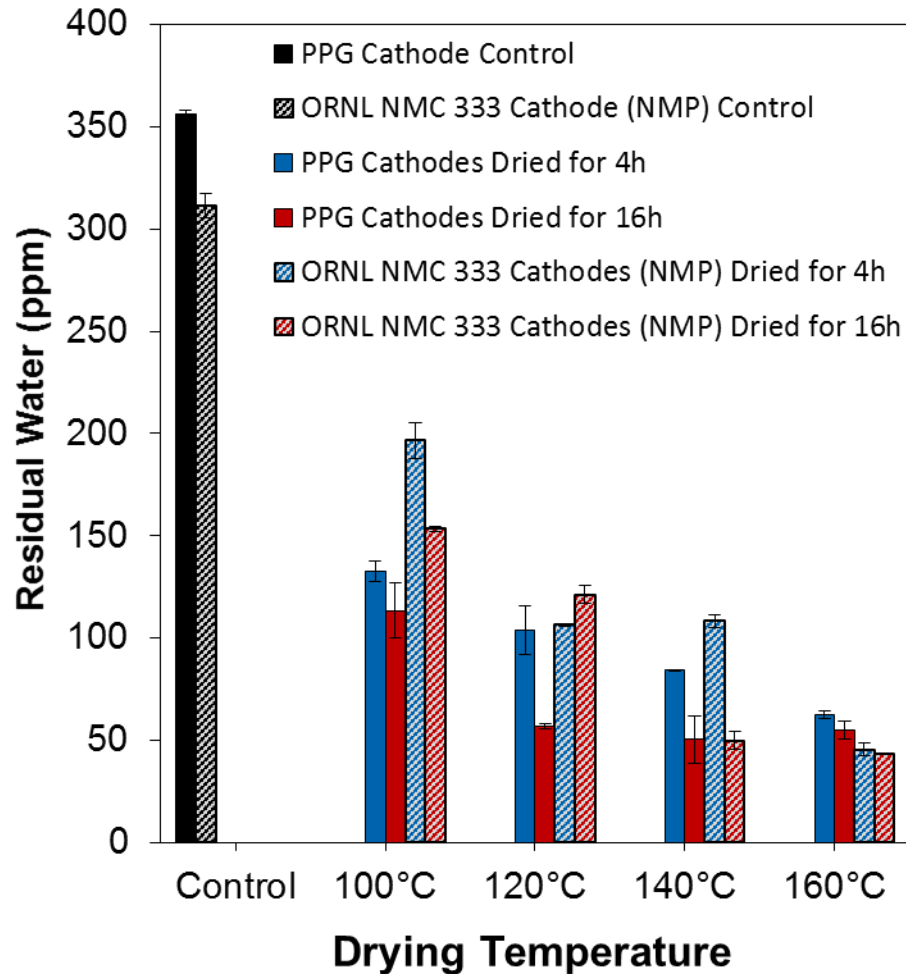
Film uniformity is controlled through particle size and deposition conditions.



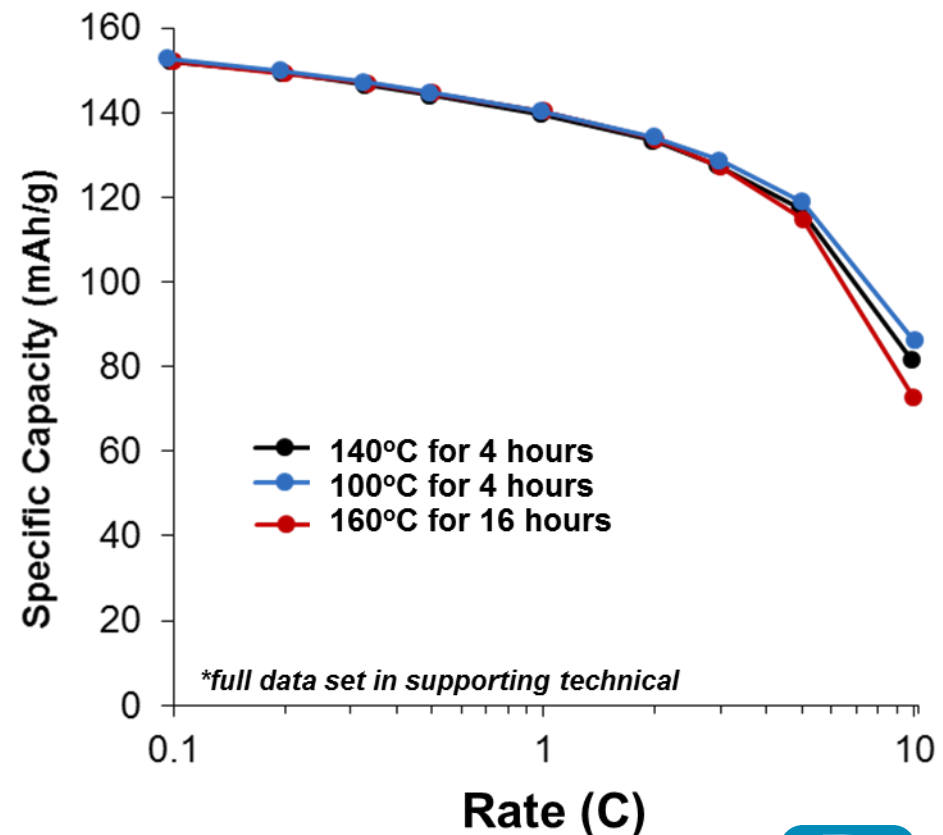


### 3. Technical Accomplishments: Optimized Drying Conditions

Water retained in electrocoated cathodes is effectively removed through secondary drying.

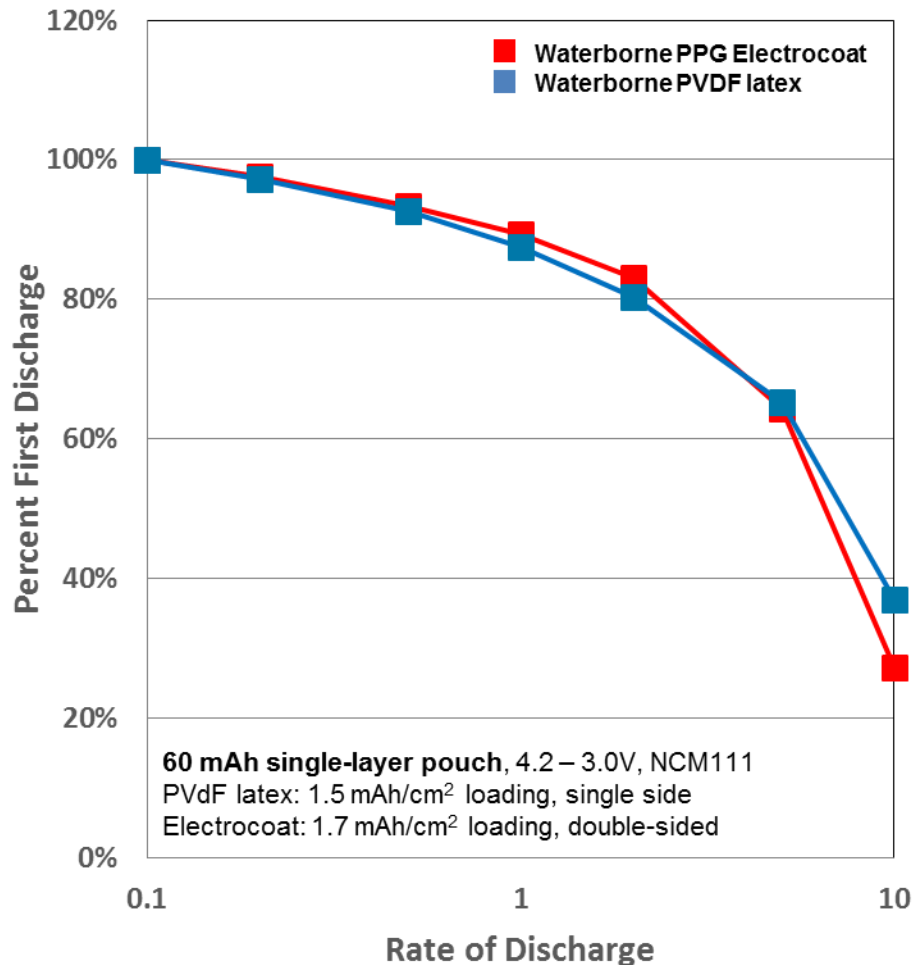


Drying conditions may impact the performance at high C-rates.

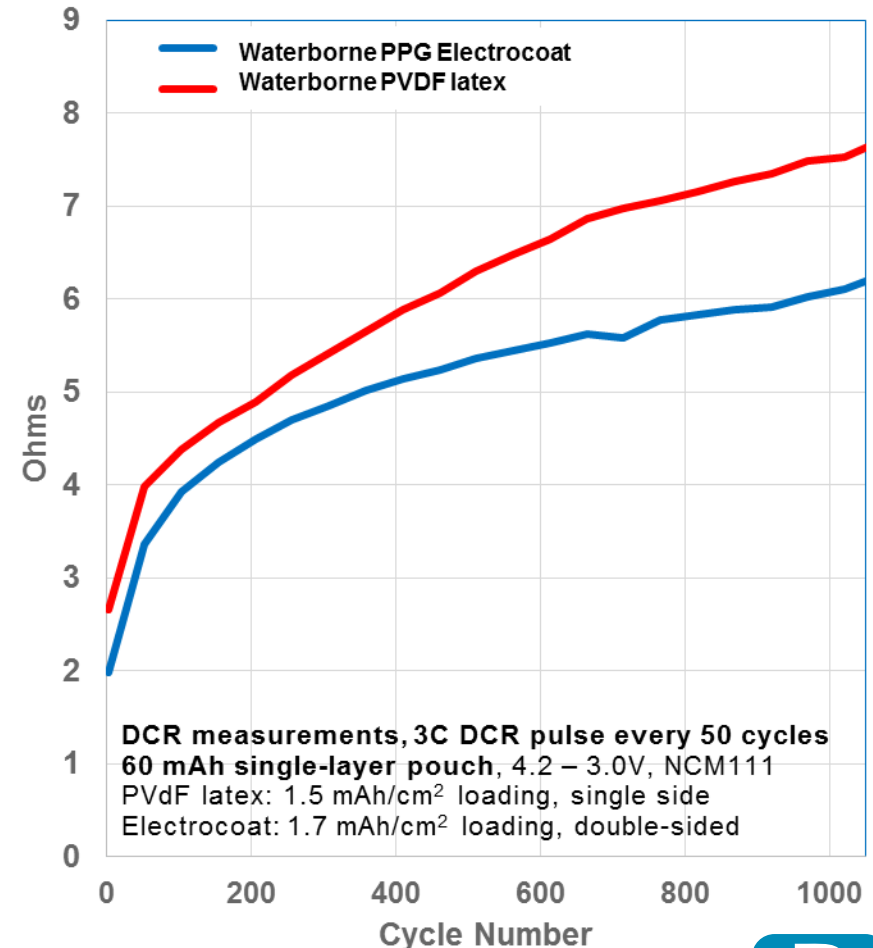


## 4. Technical Accomplishments: Pouch Cell Testing

Electrocoated NCM111 cathodes have similar performance to commercial waterborne cathodes.

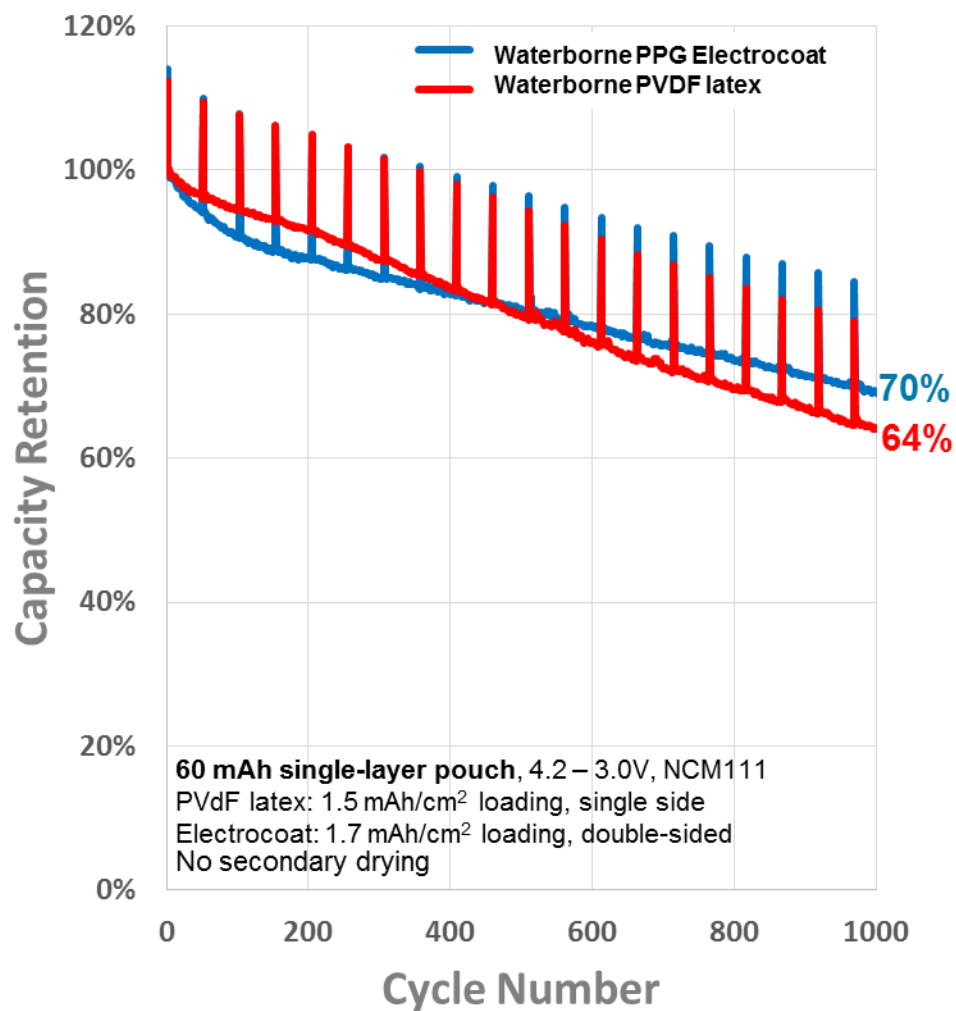


Electrocoated NCM111 cathodes exhibit lower resistance than commercial waterborne cathodes.

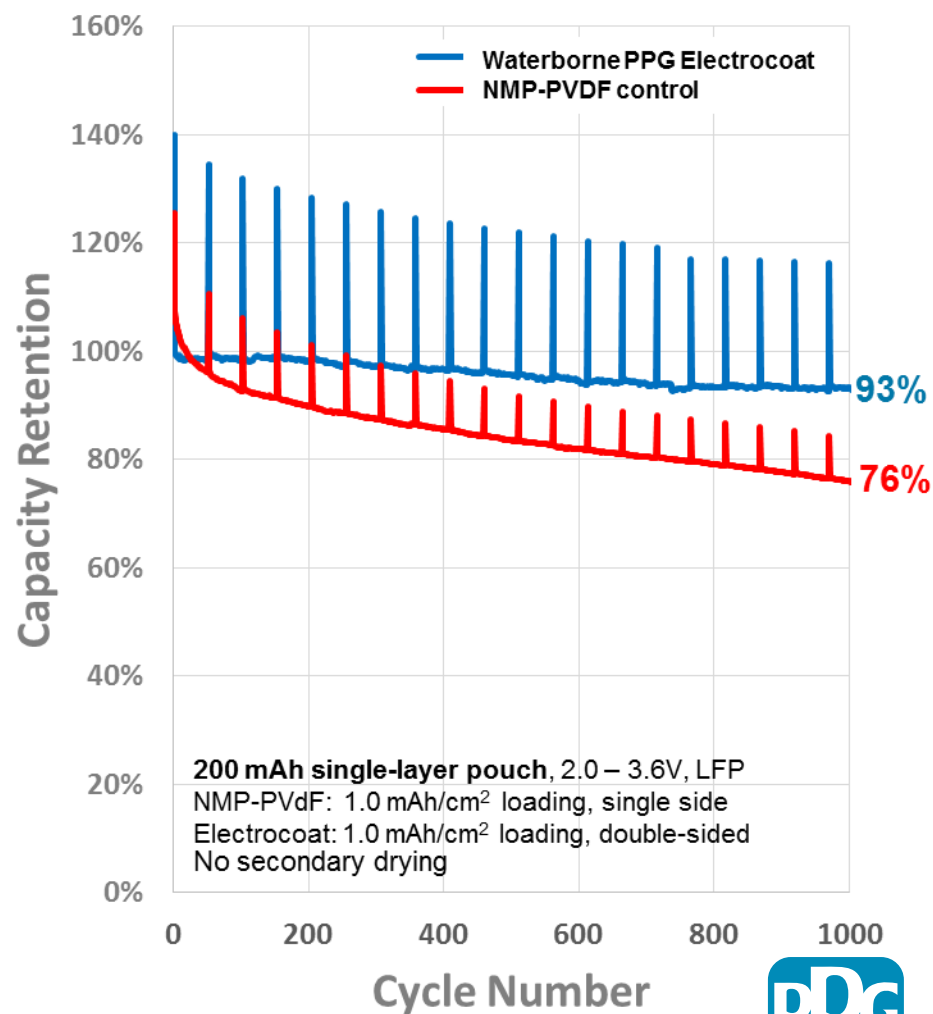


## 5. Technical Accomplishments: Pouch Cell Testing of NCM and LFP

**Electrocoated NCM111 cathodes outperform commercial waterborne cathodes at high rates.**

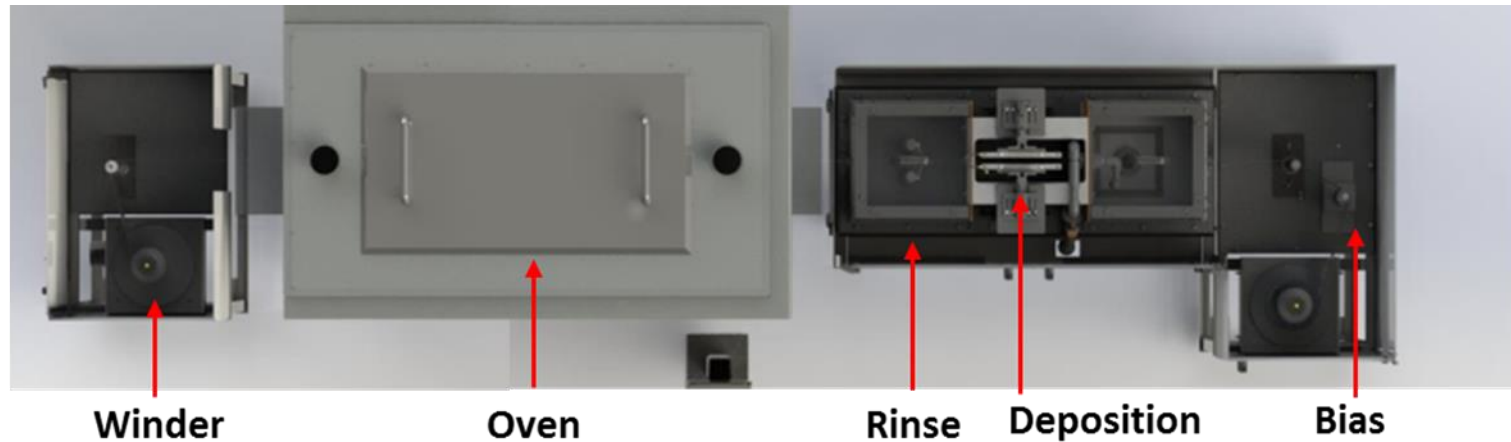


**Electrocoated LFP electrodes outperform baseline cells prepared using NMP-PVDF**



## 6. Technical Accomplishments: Pilot coater Design and Build

Pilot coater constructed and evaluated at PPG Coating Services using NCM111 active material and demonstrated continuous operation for electrocoat fabrication.



## 7. Technical Accomplishments: Mini-coater Design and Build

Pilot unit coated large sections of foil with a double-sided film with properties that matched lab-scale.

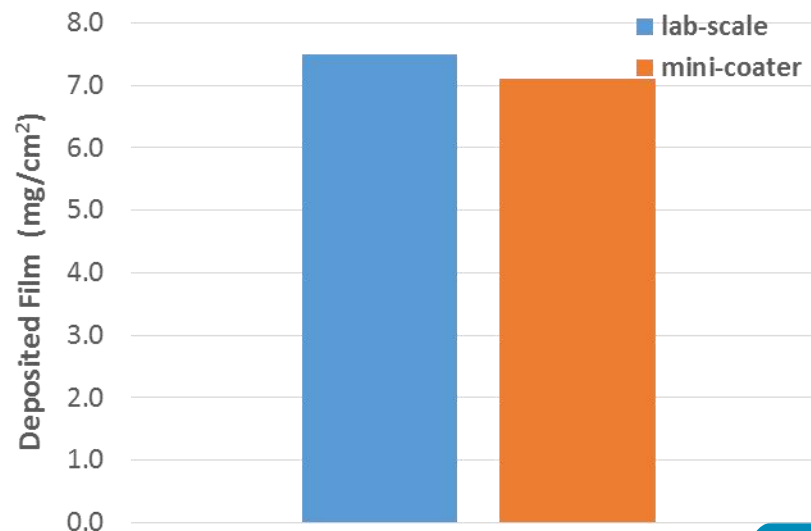
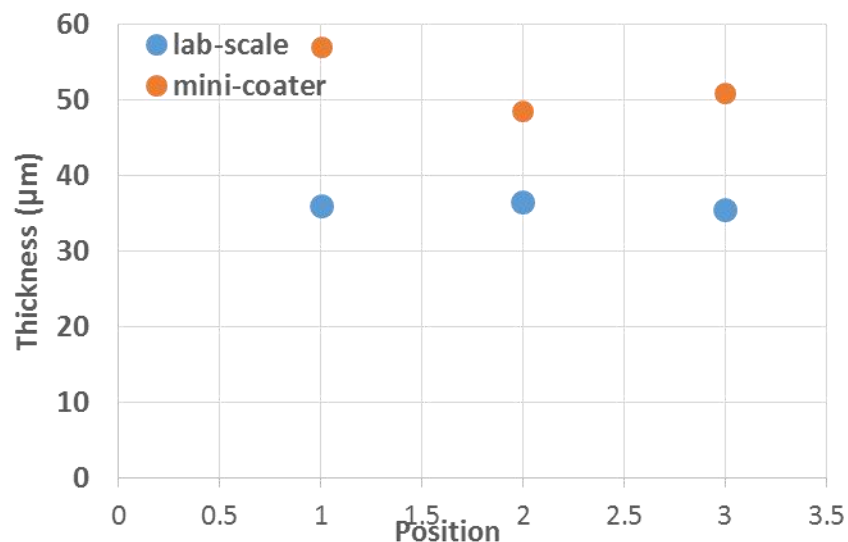
Shake-down of Pilot System Using Commercial Electrocoat Product: 8 feet



Battery: 5 feet



NCM111  
1.2 mAh/cm<sup>2</sup>



## 8. Technical Accomplishments: Cost Estimates

**BatPac estimates of 1.1% cost savings by switching to water from NMP.\***

NMC622-Graphite	NMP	Water
Cathode Solvent	NMP	Water
Solvent Used, MT/year	18,400	11,000
Solvent Recovery, %	99.5	0
Cathode Solvent Cost, K\$/year	336*	1.68
Solvent Recovery		
Capital Equipment (Installed), M\$	17.9	-
Labor, K-hours/year	16.1	-
Plant Area, m <sup>2</sup>	595	-
Plant Total		
CapEx (Installed), M\$	240	222
Labor, K-hours/year	902	886
Plant Area, m <sup>2</sup>	24,500	24,500
Pack Cost to OEM, \$/pack	\$9,619	\$9,539
Cell cost to OEM \$/cell	\$46.67	\$46.19
Calendaring \$/cell	\$0.12	\$0.12
Savings per cell, \$/cell (%)		\$0.60 (1.3%)

*\*Estimates made on a plant producing 100,000 80 kWh battery packs*

**Updated energy model from ANL predicts a savings of 1.9% from energy cost using E-coat.**


Cost	NMP	Water
Days of Operation	300	300
Solvent Needed, kg/year	19.3 M	17.7
Solvent Recovered and Recycled	94.9%	-
Thermal Energy, GWh/year	147	25
Air Heater, GWh/year	126	25
Desorb NMP GWh/year	8	-
Distillation, GWh/year	13	-
Electric Energy, GWh/year	116	12
Chiller / Condenser	39	-
Main Blower, GWh/year	68	12
2 <sup>nd</sup> Blower, GWh/year	8	-
Total Energy, GWh/year	263	37
(Drying+NMP Recovery Energy Need) : (Plant Pack Capacity) GWh/GWh)	33	5
Contribution to Pack Cost, \$/pack	\$189	\$39
Contribution to Cell Cost, \$/cell	\$1.12	\$0.23
Energy savings per cell, \$/cell		\$0.89 (1.9%)




# Response to Reviewer Comments

## Comments from 2017 AMR

“The reviewer would appreciate more focus on demonstrating good cycling and rate capability for extended time. While the reviewer understands that this is still part of the work plan so may be a non-issue, this person stated it would be nice to know if it's an issue as soon as possible.”



“It is still unclear if production rate even with double sided coating will be fast enough to overcome the slow nature of the electrocoat process. This reviewer understands that it is “tunable” but there has to be an upper bound for coating speed, and this person cannot judge if that is acceptably fast or not.”



“If anode electrocoating is planned, this person pondered whether this would be easier than cathode considering no Li leaching, or whether there is a reason a materials anode electrocoat process would not work.”

## Response to Comments

### Actions Taken

- Cell testing past 1000 cycles has now been demonstrated using single-layer pouch cells. These tests demonstrate improved stability for electrocoat cells compared to baseline cells using both NCM111 and LFP.
- Improved rate capability and lower internal resistance has been demonstrated for NCM111 cathodes in.

### Future Considerations

- Although improved performance was demonstrated for NCM111 and LFP active materials, future applications will require the use of higher energy active materials such as high-nickel NCM. One approach to enable these active materials is the use of surface treatments such as those identified for NCM622.

### Actions Taken

- Optimized electrocoat at the batch bench-scale produced a coating speed of 0.25mAh/cm<sup>2</sup>/s. If we use as an example a commercial line that applies 3.2 mAh/cm<sup>2</sup> at a linear speed of 0.17 m/s, a similar line speed and areal density could be achieved on an electrocoat line with a bath length of only about 2.2 meters.

### Future Considerations

- Optimizing film deposition rate through electrocoat deposition parameters on the pilot scale coater are expected to significantly improve the deposition rate. In particular, adjustment of the current density through counter electrode separation is an established approach to improve deposition rate.

### Actions Taken

- Preliminary experiments confirm that the electrocoating of graphite anode materials is viable with electrocoat. Unlike electrocoat of cathode materials, the pH of graphite solutions was easily controlled which will improve the compatibility of the process with the current collector.

### Future Considerations

- Anode electrocoat is ongoing but outside the scope of this grant. This work will require stabilizing copper current collectors under an anodic potential while simultaneously electrocoating graphite material. Additional compositional changes may need to be made to the electrocoat resin to ensure chemical stability when operated as an anode binder.

# Collaboration with Others



Argonne national labs has extensive expertise in active material synthesis and characterization. This expertise was used to prepare cathode active materials suitable for electrocoat. Both particle size and particle-coating technologies were explored.



Oak Ridge National Lab has extensive expertise in waterborne cathode coating technology. This expertise was used to address challenges specific to cathodes produced by waterborne electrocoat systems. Unique drying conditions for waterborne cathodes were identified.



Navitas brings commercial insight with extensive experience implementing novel battery technologies and expertise in cell design. This expertise is being used to produce and test cells from cathodes made by electrocoat, and make substantive comparisons against state-of-the-art cathode technologies.

# Remaining Challenges and Barriers

## Active material

- Robust compatibility of electrocoat with nickel-based active materials remains a challenge.
- Applicability of surface treatments to multiple vendors and nickel content requires further investigation.
- Partnerships with active material suppliers for stable active material performance needs to be explored.



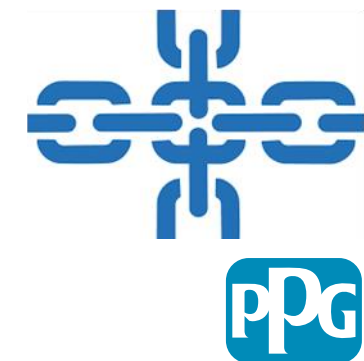
## Pilot scale production

- Maintaining uniformity, edge coverage, and areal density during continuous coating needs to be verified in the next budget period.
- Using electrocoat application parameters to control the edge profile of the coating needs to be explored in the next budget period.



## Supply chain

- Supplying a fully formulated solution is disruptive to the current battery supply chain.



# Future Work

FY18

## Optimize coating parameters on the pilot scale coater:

- Deposition conditions including applied voltage and current need to be adjusted to produce sufficient thickness during continuous movement of the coated foil.
- Oven baking conditions will be refined to produce coated foils that can be rolled up without damaging the coating

## Produce large format ( > 1 Ah) cells from foils coated by the mini-coater:

- Initial screening of pilot scale coater conditions will be executed using > 1 Ah double layer pouch cells with a loading in the range of 1 – 3 mAh/cm<sup>2</sup>.
- Optimized coater conditions will be used to produce 47 prismatic cells >1 Ah and compared with 12 baseline cathodes
- Failure analysis will identify root cause failure mechanisms.

Pilot coater installed at PPG facility



FY19

## Complete cell testing:

- Cell testing will be completed at the beginning of FY19 and 18 cells will be delivered to the DOE for evaluation

## Project completion.

1.3 Ah jelly roll w/ electrocoat cathode



# Summary



**Commercially available active materials are stable in the electrocoat system and are coated with acceptable uniformity and energy density.**

- Electrocoated cathodes demonstrated improved performance compared to cells fabricated using traditional NMP-PVDF processing methods or other commercial waterborne binder systems.



**Coating system parameters optimized to rapidly produce uniform electrodes.**

- Electrodes with acceptable uniformity and areal density were produced in a batch process to fabricate pouch cells.



**Pilot scale mini-coater construction is complete and up to 8 feet of coated foil has been produced.**

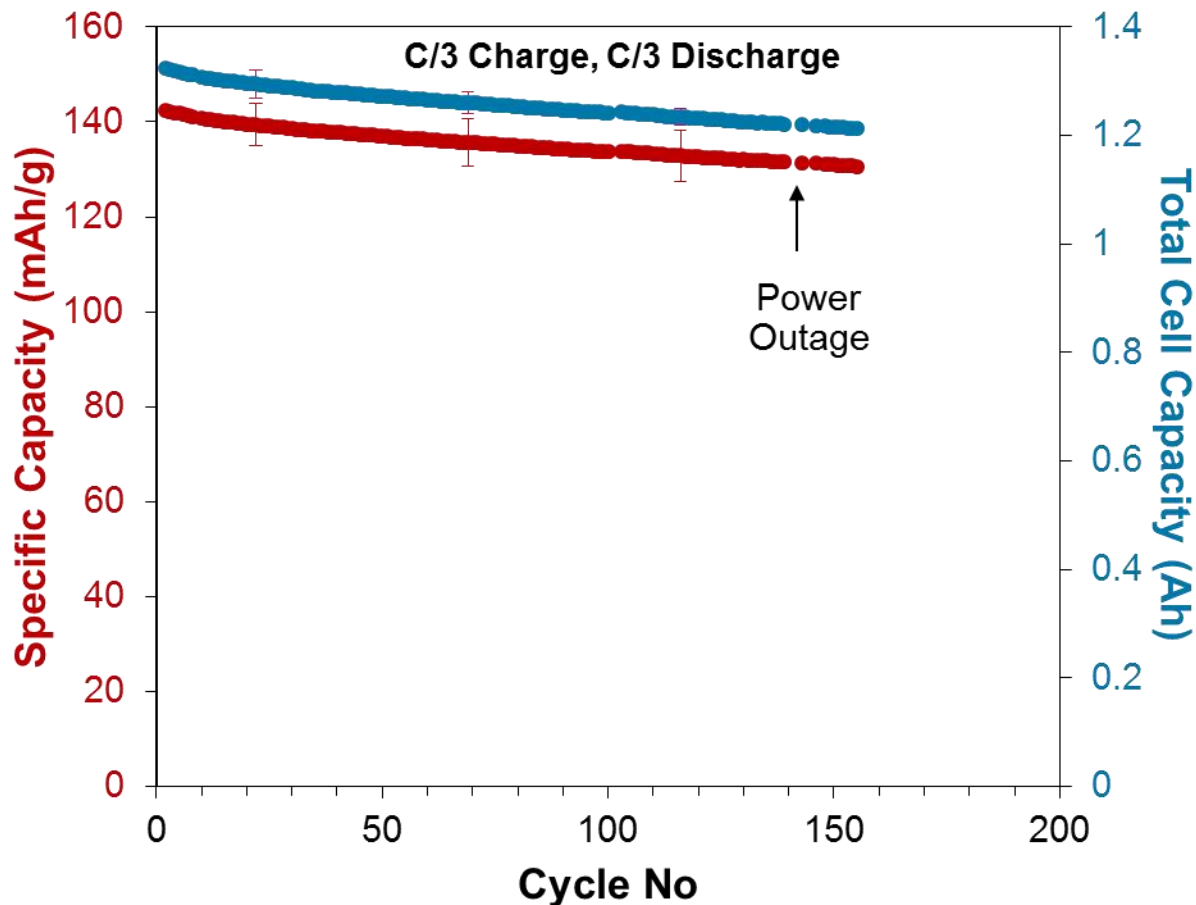
- Coatings produced on the mini-coater possess uniformity and areal density comparable to coated foils produced in a batch process.

## **Technical Back-up Slides (None)**



# 1. Technical Backup: 1.3 Ah Pouch Cell Performance

1.3 Ah Pouch cells fabricated using electrocoated NCM111 exhibit acceptable cycle life over 150 cycles.



Completed 1.3 Ah Pouch Cell

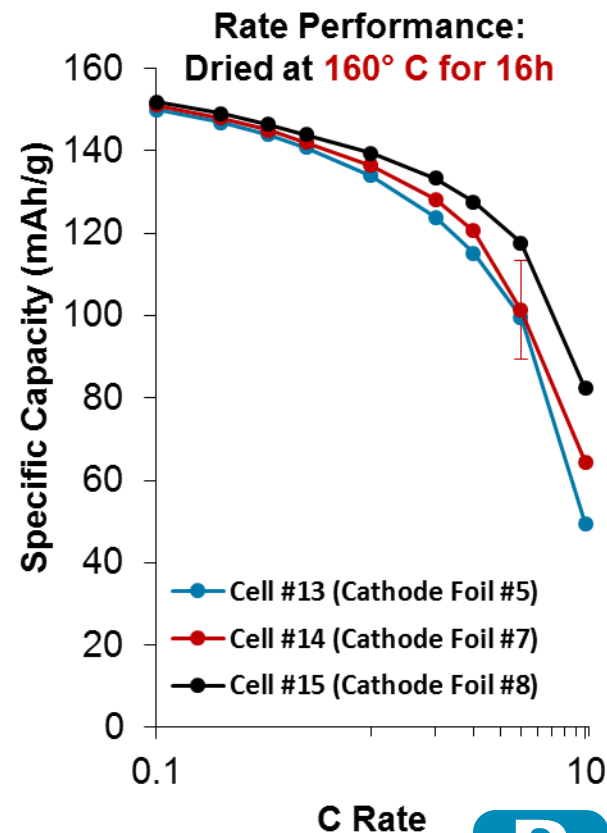
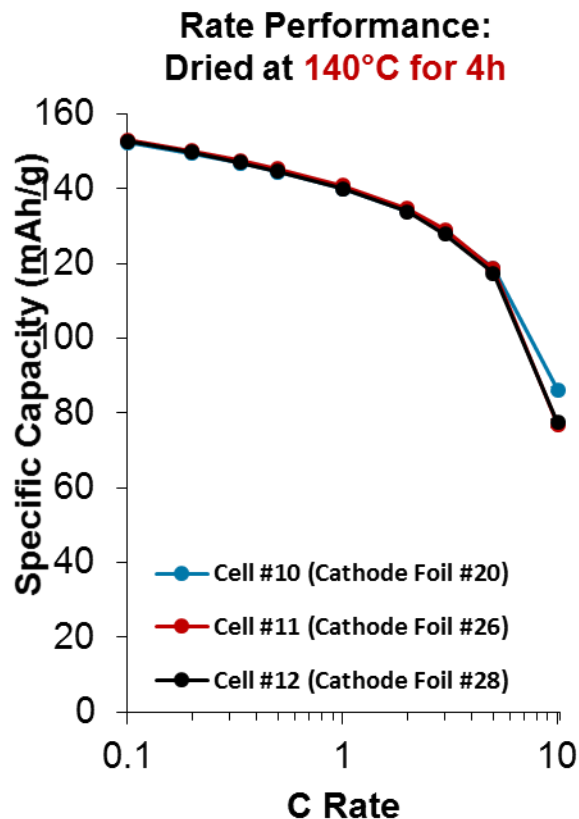
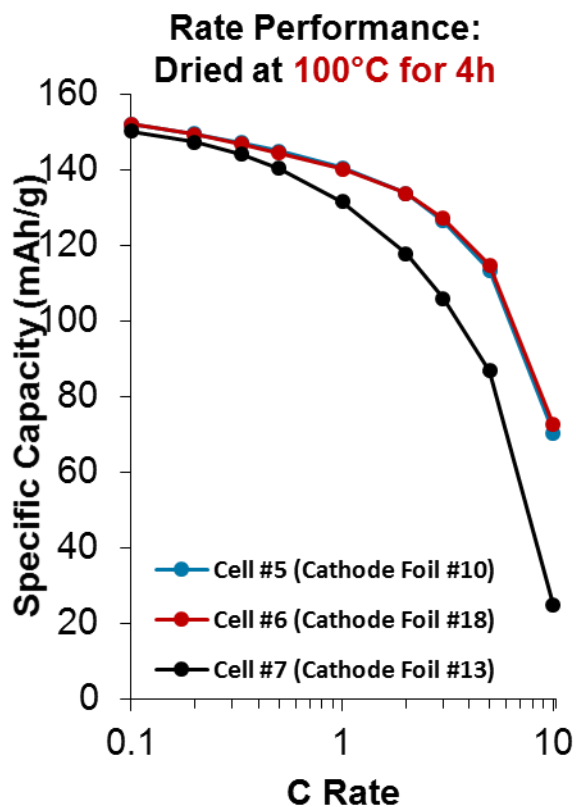


Side View of Jelly Roll

## 2. Technical Backup: Temperature Study Cell-to-Cell Variation

Cell-to-cell variation in rate performance was observed during optimization of the drying conditions.

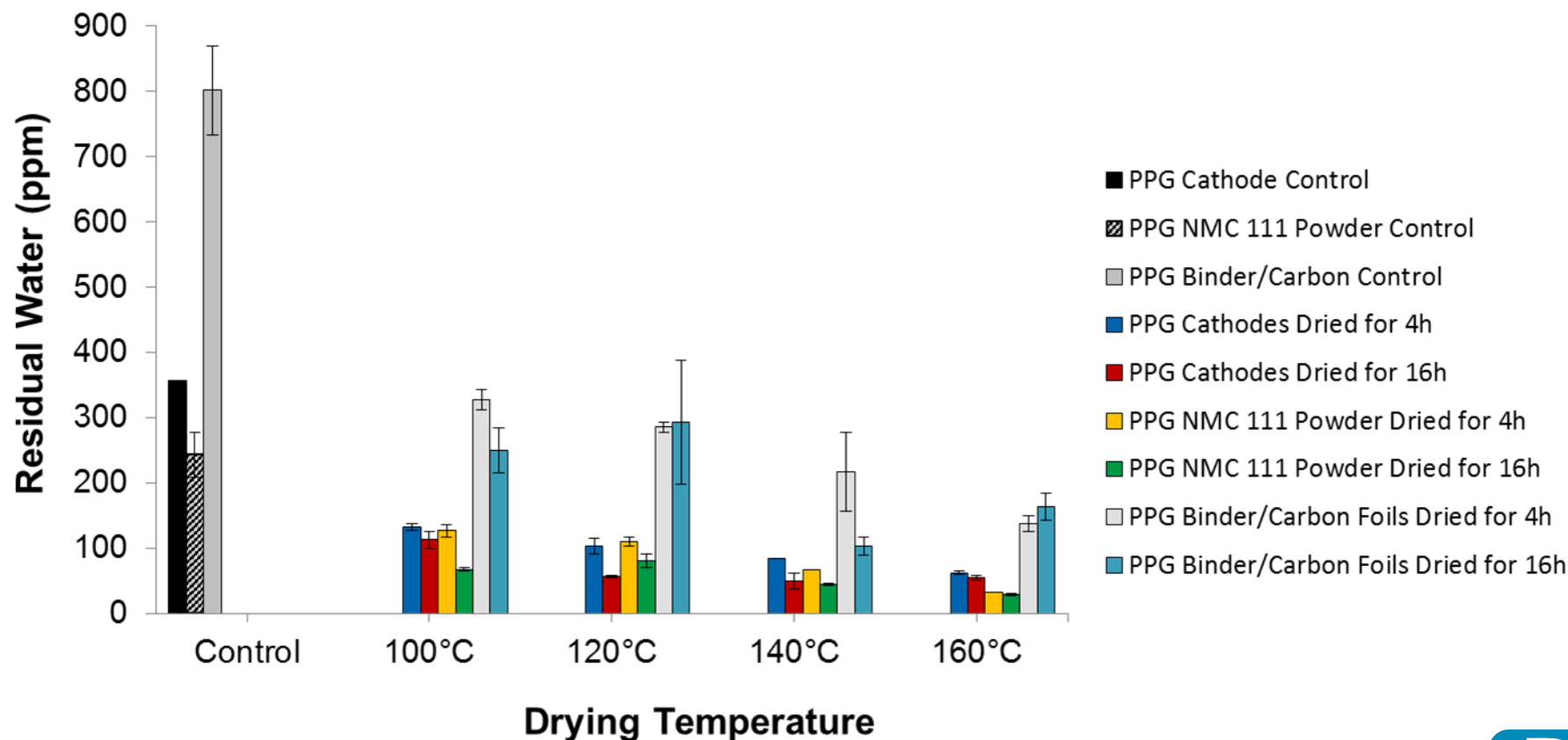
Batch-scale, bench-top fabrication of electrocoated electrodes contributed to cell-to-cell variation.



### 3. Technical Backup: Drying of Individual Components

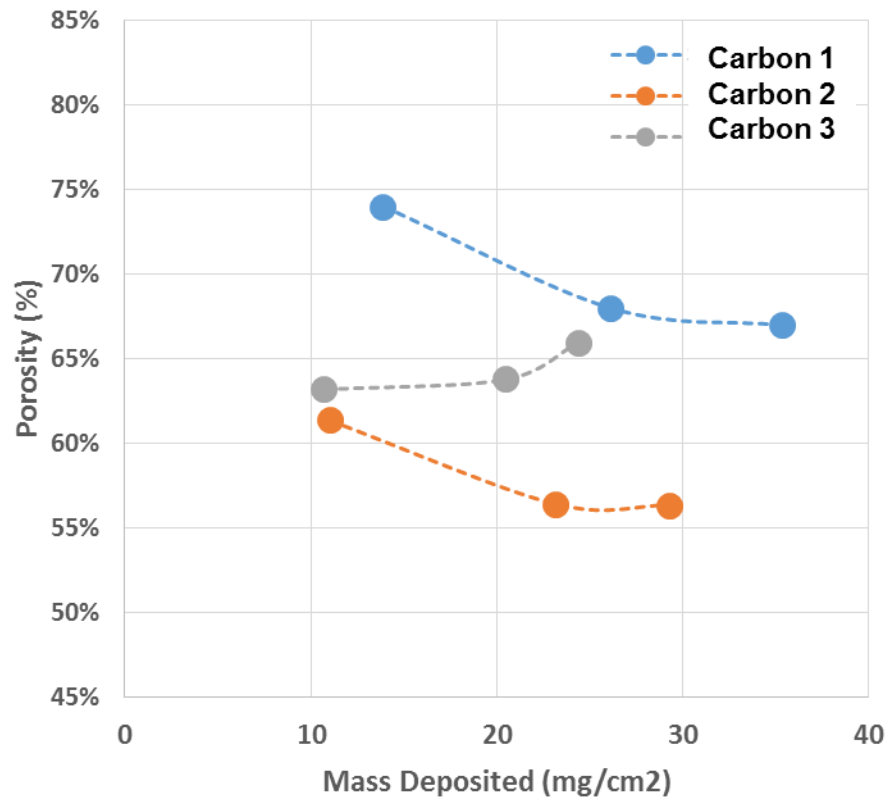
**Binder and carbon retain significantly more water than NCM111 materials.**

**PPG Cathode Components: KF Titration Results**



## 4. Technical Backup: Controlling Film Porosity

**Carbon materials have a significant impact on electrocoat film porosity.**



**Porosity may be tuned via de-watering of the electrocoated film.**

